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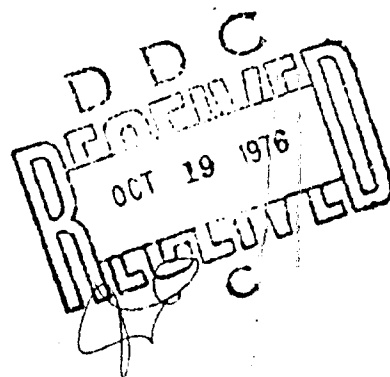
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NRL Memorandum Report 3359

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**Adsorption and Condensation of
Water on Mirror and Lens Surfaces**

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September 1976



This work was sponsored by the High Energy Laser Project Office
PM-22/PMS-405 of the Naval Sea Systems Command.



NAVAL RESEARCH LABORATORY
Washington, D.C.

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER NRL Memorandum Report 3359	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) ADSORPTION AND CONDENSATION OF WATER ON MIRROR AND LENS SURFACES.	5. FUNDING NUMBERS & PERIOD COVERED Final report, one phase of a continuing NRL problem.	
7. AUTHOR(s) Joseph A. Curcio	6. PERFORMING ORG. REPORT NUMBER	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Naval Research Laboratory Washington, D.C. 20375	8. CONTRACT OR GRANT NUMBER(s)	
11. CONTROLLING OFFICE NAME AND ADDRESS Naval Sea Systems Command (PM-22/PMS-405) Washington, D.C. 20362	10. PROGRAM ELEMENT, PROJECT, TASK NRL Problem R65-31C	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	12. REPORT DATE September 1976	
	13. NUMBER OF PAGES 20	
	15. SECURITY CLASS. (of this report) UNCLASSIFIED	
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited. 14 NRL-MR-3359		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Water adsorption Water condensation Aerosol scattering Mirror scatter		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) This is a report of a literature search on adsorption and condensation of water on mirror and lens surfaces. Adsorbed water films on clean surfaces are generally < 3.0 nm thick and can be ignored when considering absorption losses. When the surfaces are exposed to ambient conditions, various aerosols of dusts and hygroscopic particles are deposited on the surfaces which give rise to small losses by scattering of the incident radiation. In the case of hygroscopic particles the problem can be especially troublesome because here the scattering magnitude is a function of the relative (Continues)		

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20. Abstract (Continued)

humidity. For a given aerosol deposit an optical system containing a 6 mirror train will have a cumulative reflectivity decrease of about 7% as the relative humidity varies from 60% to 80%.

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A

ADSORPTION AND CONDENSATION OF WATER ON MIRROR AND LENS SURFACES

INTRODUCTION

In all field work dealing with the study of the propagation of optical radiation through the lower atmosphere, one is concerned with mobile based optical instrumentation having lens and mirror components. In the outdoor environment these lenses and mirrors are exposed to various conditions of temperatures and humidity and careful precautions must be taken to protect the surfaces from dust and also adsorption and condensation of water vapor. At low humidity locations where the air is clean it is generally sufficient to have the optical elements loosely jacketed with plastic covers. At other locations for example near a shore line where the humidity is high, the atmosphere may be contaminated with salt spray and careful precautions must be taken at all times to prevent moisture and spray accumulation on the surfaces. The spray and dust particles on the optical surfaces will scatter radiation and will introduce serious errors in experimental spectral extinction data.

Adsorption of Water on Solid Surfaces

The adsorption of water on solid surfaces has been studied by many workers. Water film thickness on various glasses determined by Veith⁽¹⁾ is shown in Fig. 1. There is a great deal of work on adsorption of water on glass some of which indicates adsorbed films up to $0.1\mu\text{m}$ in thickness. A careful experiment by Bowden and Throssell² shows that the adsorption of water on a clean gold surface at water vapor pressure near saturation corresponds only to about 2 molecular layers. These experiments suggest that the thick adsorption layers reported by other workers is due to surface contamination by dusty hygroscopic particles and other aerosols that act as condensation centers.

In an optical study of adsorbed water films by Frazer³, it was found that the water film thickness on a clean glass surface remained constant at about 1 mono molecular layer to about 6 mm H₂O vapor pressure. From then on there is a cluster formation which results in a gradual covering of the surface until at 12.5 mm pressure there is another monomolecular layer present. Above 12.5 mm pressure there is strong condensation so that by 16 mm pressure (89% RH) the adsorbed layer has a thickness of about 3.0 nm.

Note: Manuscript submitted August 25, 1976.

Multilayer adsorption of water near saturation pressure (99.18% RH) was studied by Garbatski and Falmon⁴. Their results show a minimum film thickness of 62.0 nm near the saturation pressure. They conclude that multilayer adsorption is a physical fact and that there may be established a continuous transition from adsorption to condensation at high vapor pressures.

Work by Davies⁵ on incipient condensation of water vapor on a gold surface shows measurable scattered visible light from the surface exposed to room air at a relative humidity of about 85%. The scattered signal increased by an order of magnitude at a relative humidity of about 98%. This is about 20X greater than the scattered visible light from the same surface at the same relative vapor pressure after prolonged washing. The increase in signal being attributed to water soluble aerosols in room air. The mechanism of incipient condensation appears to be governed largely by the effects of soluble impurities.

Davies assumes that the scattering is caused by discrete liquid particles (droplets) lenticular in shape and uniform in size. These droplets behave as independent scattering centers so that the scattering by N droplets is N times that for an individual droplet and that they scatter light in the same way as isolated spheres of the same radius of curvature. The lenticular form of constant shape is consistent with the view that the surface possesses a very fine structure of molecular dimensions and a much coarser structure due to polishing. The droplet being intermediate in scale between the two.

It is apparent then that heavy deposition of water will occur only in the presence of contamination on the surface. In the carefully controlled clean surface experiments by Bowden and Throssell, the observed film thickness did not exceed 30 nm near saturation conditions and not much more than 2 molecular layers thick from unsaturated vapor. Other workers in ordinary laboratory environment have shown that water vapor films as thick as 0.10 μm can be deposited on a surface from a near saturated vapor. It should be noted that the large, so-called film thickness was an equivalent thickness determined from area and weight considerations. In reality the condensation is clustered about the aerosol deposits which have diameters many times the quoted average thickness.

The possibility of obtaining the infrared absorption spectrum of a layer of material situated on a metal surface by reflections from the surface has been investigated by Greenler⁶. He concludes that there is little interaction between an electromagnetic wave and a thin film such as an adsorbed layer 3.0 nm thick. Burch⁷ has conducted experiments on absorption by H₂O films adsorbed on mirrors. His carefully controlled experiments indicate that there was no detectable cumulative absorption from as much as 36 near-normal multiple reflections from a mirror with an adsorbed H₂O film. Pickering⁸ attempted to obtain spectra of molecules adsorbed on metal films. With 35 multiple reflections at near normal incidence he observed weak absorption structure which he

attributed to water vapor in the absorption cell. From the above it is clear that ordinary adsorbed water films on the mirrors will be of no consequence in the extinction experiments.

Aerosol Deposits on Surfaces

The field operations which have been carried out near the ocean beach at Cape Kennedy, Fla., have been in an environment of generally high humidity and in an atmosphere highly contaminated with salt water aerosols of various sizes which originated from the wave spray along the beach. The receiver and transmitter trailers were placed near the beach which made the optical elements within vulnerable to the formation of water deposits on their surfaces. At the receiver there were 5 mirror surfaces plus the integrator and the detector window. The transmitter system had as many as 20 surfaces plus the integrator and the detector window. Normally when not in operation the surfaces were covered with loose fitting plastic jackets. The trailer doors are closed and the interior is heated above the dew point so that the surfaces are reasonably well protected. However, during data runs the large trailer doors are open and the interior is at ambient conditions. The interior must be at ambient temperature to minimize turbulence effects from temperature gradients at the aperture. These conditions may last as long as 5-6 hours. For 1 days' operation during which time aerosols may deposit on the more vulnerable surfaces located near the outgoing and incoming apertures. Surfaces recessed within the trailers are less vulnerable than those near the large doors and the side door. From the configuration of the optical systems in the trailers it is estimated that only 1/3 of the transmitter mirrors were fully exposed to the ambient and only 1/2 of the receiver mirrors were similarly exposed. Fig. 2 is a 45x photo of a small area (.175"x.225") of the turning mirror located near the aperture of the receiver trailer taken at NRL at the conclusion of the Fla. tests in 1975. An approximate densitometric analysis shows that about 40 percent of the surface is covered with contaminants.

The question now arises: how does this contamination affect quantitative extinction data? Obviously the aerosol deposit will scatter out radiation and may introduce errors in the extinction coefficient results. When we consider the many surfaces in the receiver and transmitter and possible aerosol deposits then become a serious problem. Some of the error will cancel out since data reduction involves a ratio of receiver to transmitter signals. At the present time we use a zero run technique to calibrate out the losses at the optical surfaces. These zero runs are made periodically and in principle the scattering effect from the cumulative aerosol deposit on the various surfaces should cancel out. The degradation of the calibration factors between zero runs is assumed to be some function of the time period between zero runs.

There is a potentially serious problem with this technique and it has to do with the nature of the aerosol deposit. Being located near the ocean shoreline the aerosol is predominantly hygroscopic because of its sea water origin. During conditions of strong offshore breezes the

aerosol will contain a substantial hygroscopic component. One of the properties of these hygroscopic particles is that the particle size is a function of the relative humidity. Continental aerosol particles are also sensitive to relative humidity. It is this factor which is critical to the calibration procedures in these experiments.

Growth of Hygroscopic and Insoluble Particles

The growth of hygroscopic and insoluble particles as a function of relative humidity has been studied by many workers, G. Hanel⁹, P. Winkler¹⁰, C. Orr¹¹, Rozenberg¹². Work by Orr et al in 1958 shows that the behavior of hygroscopic particles exposed to increasing humidity is to adsorb moisture amounting to a few molecular layers at low relative humidity; to dissolve as the relative humidity increases, becoming saturated droplets and at the same time undergoing an abrupt size increase and thereafter as relative humidity increases still further to grow larger and more dilute. The particle decreases in size as the relative humidity is decreased although a hysteresis is noted because the particle is recrystallized at a relative humidity much lower than the saturation relative humidity noted on the growth portion of the curve. This is illustrated in Fig. 3 which shows size of hygroscopic particles as a function of relative humidity. Studies by Orr and Twomey indicated that the transition from particle to droplet occurs at higher and higher relative humidities as the initial particles size is increased. At about 75% relative humidity all size particles are in solution as droplets. The hygroscopic aerosol in the atmosphere has a broad range of particle size and does not show the abrupt discontinuities seen in the single particle growth curve. This is shown in Fig. 4 from Hanel. The dashed growth curve is for a mixture of sea-salt particles $> 0.04\mu\text{m}$ radius. The solid growth curve is for a continental aerosol mixture of soluble and insoluble particles measured at Mainz, Germany during summer 1966 from Hanel. Table 1 gives the ratio r/r_0 as a function of relative humidity for 5 values of r . Table 2 gives the ratio r^1/r_s as a function of relative humidity for a sea-salt aerosol where r_s is $> 0.04\mu\text{m}$. These tables and curves show the large variation in particle-droplet size as a function of relative humidity. In the case of the hygroscopic particles the average radius is increased by a factor of 2.3 when the relative humidity is varied from 60% to 90%. For a continental aerosol, which is a mixture of soluble and insoluble material, the average radius is increased by a factor of 1.6 for a similar change in relative humidity.

Scattered Radiation from Optical Surfaces

From the above, it is obvious that aerosol deposits on mirrors and lenses could modify experimental extinction data. The amount of radiation scattered from the optical surfaces will be a function of the particle number, size, refractive index and wavelength of the incident radiation. As the particle size increases with increasing relative humidity, there will be a corresponding increase in scattered radiation. This is illustrated in Fig. 5 which shows increase in

visible scattered light as a function of relative humidity in a photoelectric dew point device.⁽¹⁴⁾ Curve (a) for a deteriorated mirror exposed to laboratory air similar to a continental aerosol, shows that at 90% relative humidity the scattered light is about 5% that for a heavy dew deposit or total scattering. At 75% relative humidity the scattered signal is about 1% of the total incident signal. These figures are only estimates since no quantitative information is available for the aerosol deposit on the deteriorated dew point mirror. It is assumed that scattered radiation in the field of view solid angle of the receiver was proportional to total volume scattering and further that the signal for heavy dew deposit in Fig. 5 is proportional to 100% scattering. In the infrared at $3.8\mu\text{m}$ scattering is less effective and the scattered signal may be down by a factor of 4 giving scattering factors of 0.25% and 1.25% for deteriorated surfaces as the relative humidity varies from 75% to 90%.

For the field experiments in Florida, I have estimated 16 surfaces at the receiver and 10 at the transmitter. Using growth factors from Table 2 and scattering factors from Fig. 4 as a function of relative humidity, the cumulative reflectivity of transmitter-receiver optics as a function of relative humidity for continental and hygroscopic aerosol is shown in Table 3. Reflectivity ratios are also shown as a function of relative humidity. Table 3 also shows expected ratios when zero runs are made when the relative humidity is 60%, 70%, 80% and 90%. For both continental and hygroscopic, the ratio spread seems to be lowest when the zero runs are made at the lowest relative humidity. The important feature here is that reflectivity is an inverse function of relative humidity. Consequently data runs made when relative humidity differs from the zero run will have an error. For example, zero run made at 70% relative humidity and data run made at 80% relative humidity will have a reflectivity factor of 0.06 which may be erroneously attributed to path transmission.

To this point we have been concerned with a fixed average aerosol. In actual field work where the surfaces are exposed to ambient conditions there is a cumulative aerosol build up on the optical surfaces which must be considered. Factors to be considered in estimating the aerosol accumulation are exposure time (5 hrs./day), location of surfaces in the trailers relative to apertures, air motion in trailers which is to ambient wind speed, and aerosol concentration. It seems reasonable to assume 4 surfaces at the transmitter and 2 at the receiver to be most vulnerable to ambient aerosol accumulation. For aerosol concentration we take an experimental distribution measured at the site on day 74 when relative humidity was 72.4% and the total aerosol cross-section was $20\mu\text{m}^2/\text{cm}^3$. Assuming the following:

Air current in trailers - 1cm sec^{-1}

particle cross-section - $20\mu\text{m}^2 \text{ cm}^{-3}$

particle deposition efficiency - 10^{-1}

exposure time - 5 hrs./day

total area of particles accumulated on 1 surface at 70% RH =
 $3.6 \times 10^4 \mu\text{m}^2/\text{day}$

If the time interval between zero runs is 10 days then the accumulated particle area is $3.6 \times 10^5 \mu\text{m}^2 \text{ cm}^{-2}$ for each surface. For these estimation purposes we assume an air current velocity in the trailer of about 6 cm sec^{-1} which gives a total accumulated cross-section particle area of $2 \times 10^6 \mu\text{m}^2 \text{ cm}^{-2}$ or about 2% of the area at 70% RH which is the minimum accumulation on each surface. Assuming 6 surfaces vulnerable to the ambient aerosol gives an aerosol count equivalent to that contained in a 6 km path on day 74.

Absorption by Water Droplets

A third factor not generally considered is absorption by liquid water droplets. It is clear from the literature that the continuous adsorbed films on clean surface have thickness of only a few angstroms and are of no consequence as absorbers. In the present situation the surfaces are not scrupulously clean but are contaminated with hygroscopic particles which are randomly dispersed. The best estimate is that these particles have the same size distribution as the atmospheric aerosol and have the same scattering properties. There is a possibility that the larger particles have a lower deposition efficiency and hence the surface may be deficient in larger particles. At high relative humidity when the particles have increased in size by a factor of 2 this will not be a serious factor. At higher humidities $> 75\%$ RH the particles on the surface may be treated as an atmospheric haze aerosol. Deirmendjian⁽¹⁵⁾ states that the extinction of water droplets is not entirely the result of scattering but it includes some real absorption within the droplet. In general this absorption is not equivalent to the bulk absorption of a water layer corresponding to the mass of the water droplets. The reason for this is that in large droplets about one half of the incident radiation is diffracted without penetrating the droplet. In small droplets the extinction cross-section is mostly accounted for by absorption in the spectral region of high water absorption coefficients. The important factors in estimating liquid water absorption are relative size, size distribution, concentration and the spectral extinction coefficients. Figure 6 from Deirmendjian shows spectral extinction and some scattering coefficients for various distributions of water droplets. Curve M for a maritime aerosol shows maxima at $3.0 \mu\text{m}$ and $6.0 \mu\text{m}$ which closely follow the variations in the imaginary part of the index of refraction of water at these wavelengths. It appears that scattering in the DF region at $3.8 \mu\text{m}$ which is a region of relative weak liquid water absorption is only slightly affected by water absorption when the particle distribution is similar to haze M.

Conclusions

This preliminary literature search shows that adsorbed water films on clean mirror surfaces are generally < 3.0 nm thick and can be ignored when considering absorption losses at the mirror surface. In the propagation experiments where mirrors are exposed to the ambient, various aerosols of dust and hygroscopic particles are deposited on the surfaces. These give rise to radiation loss by scattering a small amount of incident radiation. In the case of the hygroscopic particles the problem can be especially troublesome because here the scattering magnitude is a function of the relative humidity. Table (3) shows some estimates of this effect for a given aerosol deposit where, for example, an optical system containing a 6-mirrors train will have a cumulative reflectivity decrease of about 7% as the relative humidity varies from 60% to 80%. At 90% relative humidity the loss is about 11%.

In the present propagation experiments the receiver system has a net increase of 6 mirrors over the transmitter system. In normal calibration procedures this inequality is cancelled out. In actual calibration procedures several factors must be considered. As a first thought the calibration should be carried out at relative humidity conditions similar to those during the data taking period. However since one of the goals of the experiment is to study propagation as a function of humidity, it becomes necessary to have some sort of continuous monitoring system to determine scattering losses at the mirror surfaces during the data taking period.

In this discussion we have assumed a constant aerosol deposit on the mirror surface. Experience has shown that there is an increase in the aerosol deposit on the mirror surfaces as the experiment progresses. Even in a controlled humidity-temperature environment, Hanley⁽¹⁶⁾ has observed an increase with time, in scattered signals from a power monitor mirror installed at the laser facility at CBD. Here again the monitor system will give information on the increase in aerosol deposit. To minimize variations in scattering because of aerosol composition and size distribution, the monitor should operate at the experimental wavelengths.

An important factor brought out by this literature search was the almost total lack of good quantitative data on scattered radiation by aerosol deposits on mirror surfaces. In particular what is needed is a study of the scattered signal as a function of relative humidity, and if indeed this turns out to be a serious problem for the propagation experiments, the study should include methods to monitor aerosol scattering from the mirror surfaces and also methods to make the surfaces less vulnerable to the ambient aerosol. A good recent paper on mirror scatter by Heinisch⁽¹⁷⁾ and Young was brought to the author's attention during preparation of this report. The near-axis scatter of infrared ($10.6\mu\text{m}$) energy from high reflectivity metal mirrors and the effects of contamination on scatter is described. Methods of evaluating various forms of contamination and selected quantitative data are presented.

Acknowledgements

I would like to thank P. Ulrich for his encouragement and critical review of the manuscript. Also thanks to C. Acton for the photographic work.

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Table I — Ratio of r/r_0 for a continental aerosol as a function of relative humidity for 5 values of r_0 . (Hanel)

$r_0(\mu)$	0.01	0.04	0.1	1	10
f	r/r_0				
0.3	1.04	1.04	1.04	1.04	1.04
0.6	1.17	1.17	1.17	1.17	1.17
0.8	1.34	1.40	1.41	1.43	1.43
0.9	1.51	1.66	1.70	1.72	1.72
0.95	1.68	1.93	2.01	2.05	2.06
0.975	1.79	2.29	2.48	2.58	2.59
0.99	1.86	2.79	3.19	3.45	3.48
0.995	1.89	3.13	3.83	4.30	4.36
0.998	1.91	3.48	4.69	5.81	5.91

Table II — Ratio r'/r_s as a function of relative humidity for a sea-salt aerosol where r_s is $0.04 \mu\text{m}$. (Hanel)

f	r'/r_s	f	r'/r_s
0.22	1.01	0.73	1.45
0.30	1.01	0.75	1.88
0.33	1.04	0.77	1.91
0.38	1.05	0.79	1.97
0.43	1.06	0.81	2.03
0.50	1.08	0.83	2.09
0.55	1.11	0.85	2.16
0.61	1.15	0.87	2.24
0.66	1.21	0.89	2.32
0.68	1.26	0.92	2.55
0.71	1.36	0.95	2.86

Table III — Cumulative reflectivity of transmitter receiver optics as a function of relative humidity for continental and hygroscopic aerosols

RH%	r/r_o	S(v)	S(IR)	R(IR)	<u>Continental</u>					R/R ₉₀
					[R(IR)] ⁶	R/R ₆₀	R/R ₇₀	R/R ₈₀		
60	1.17	.024	.006	.994	.964	1.00	1.005	1.015	1.037	
70	1.30	.029	.007	.993	.959	.995	1.0	1.009	1.031	
80	1.41	.034	.0085	.9915	.95	.985	.991	1.00	1.022	
90	1.70	.050	.0125	.988	.93	.965	.970	.979	1.0	
<u>Hygroscopic</u>										
60	1.14	.024	.006	.994	.965	1.00	1.013	1.076	1.13	
70	1.32	.033	.0083	.992	.953	.988	1.00	1.062	1.116	
80	2.0	.074	.0185	.982	.897	.930	.941	1.00	1.05	
90	2.4	.106	.0265	.974	.854	.885	.896	.952	1.00	

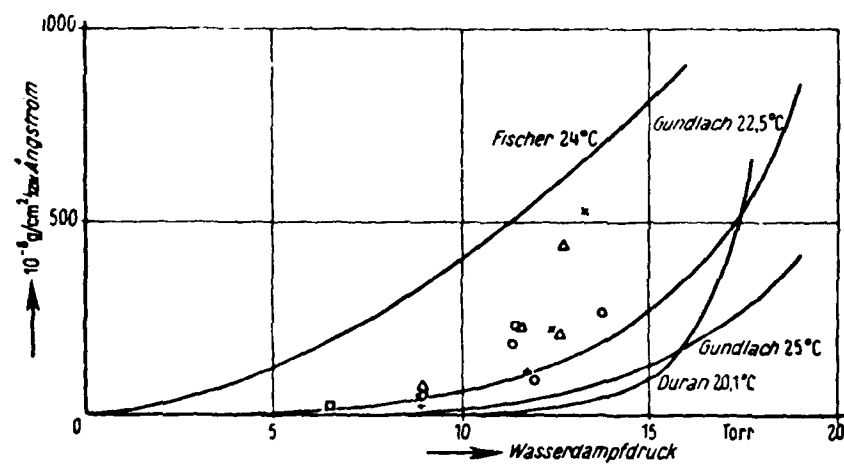


Fig. 1 — Water volume per unit area for various types of glasses and its dependence on water vapor pressure. (Veith)

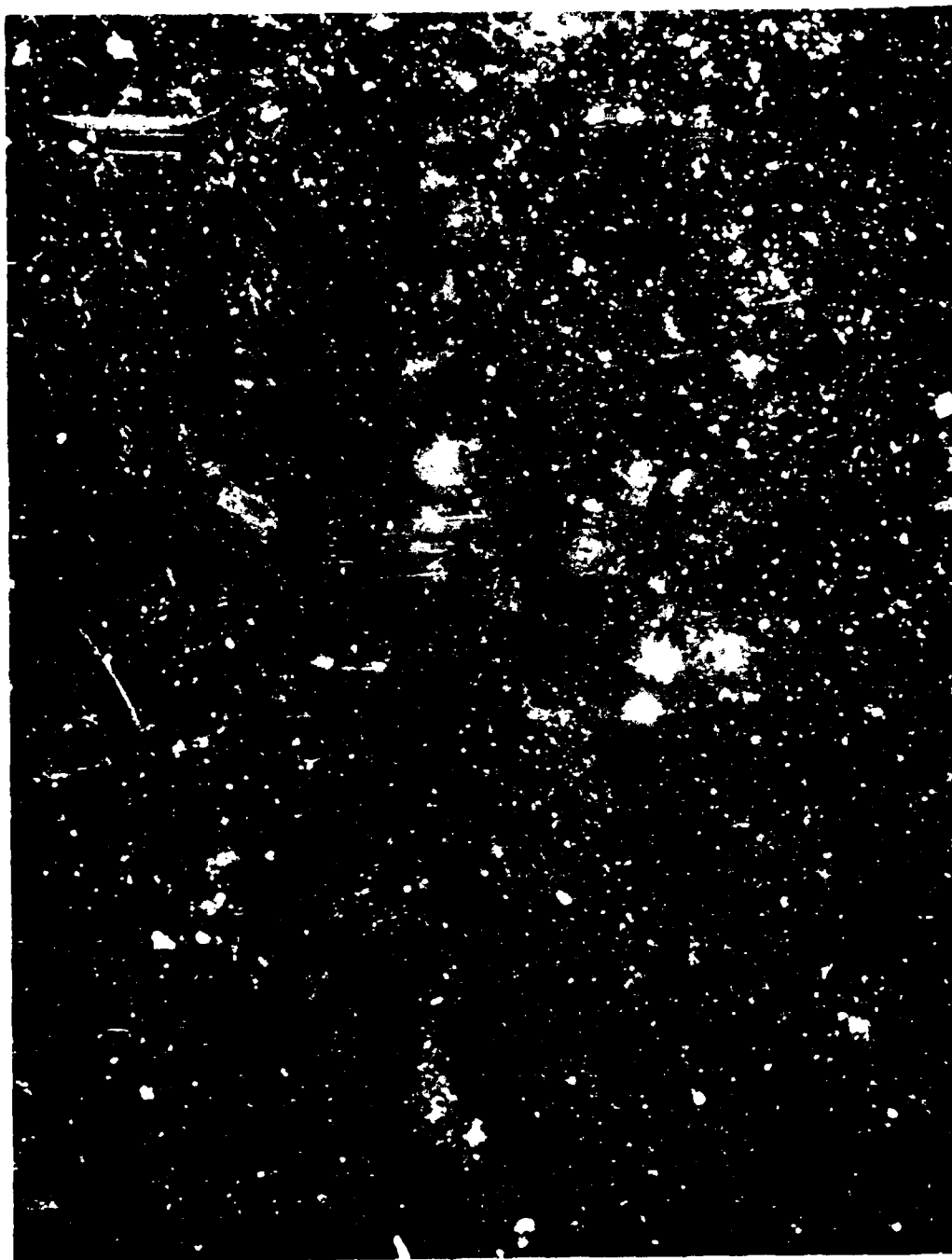


Fig. 2 — Magnified photo, 45x, of a small area of the turning mirror
located at the aperture of the receiver trailer

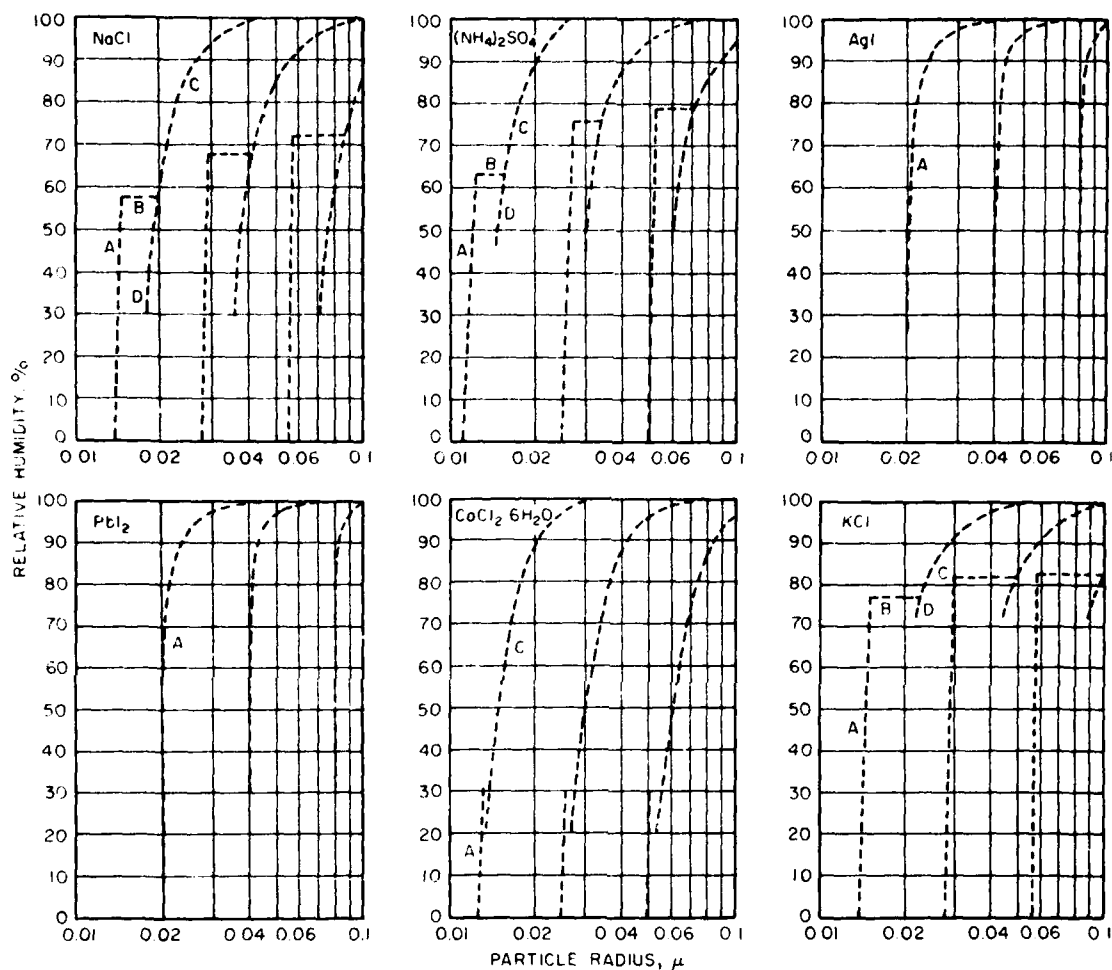


Fig. 3 — Calculated aerosol size as a function of relative humidity for several hygroscopic particles. (Orr)

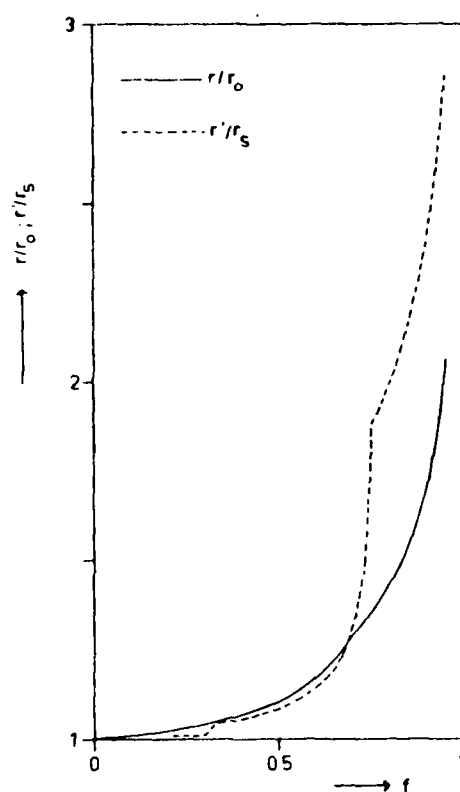


Fig. 4 — Ratio r/r_0 for continental aerosol particles and ratio r'/r_s for sea-salt particles as a function of relative humidity. (Hanel)

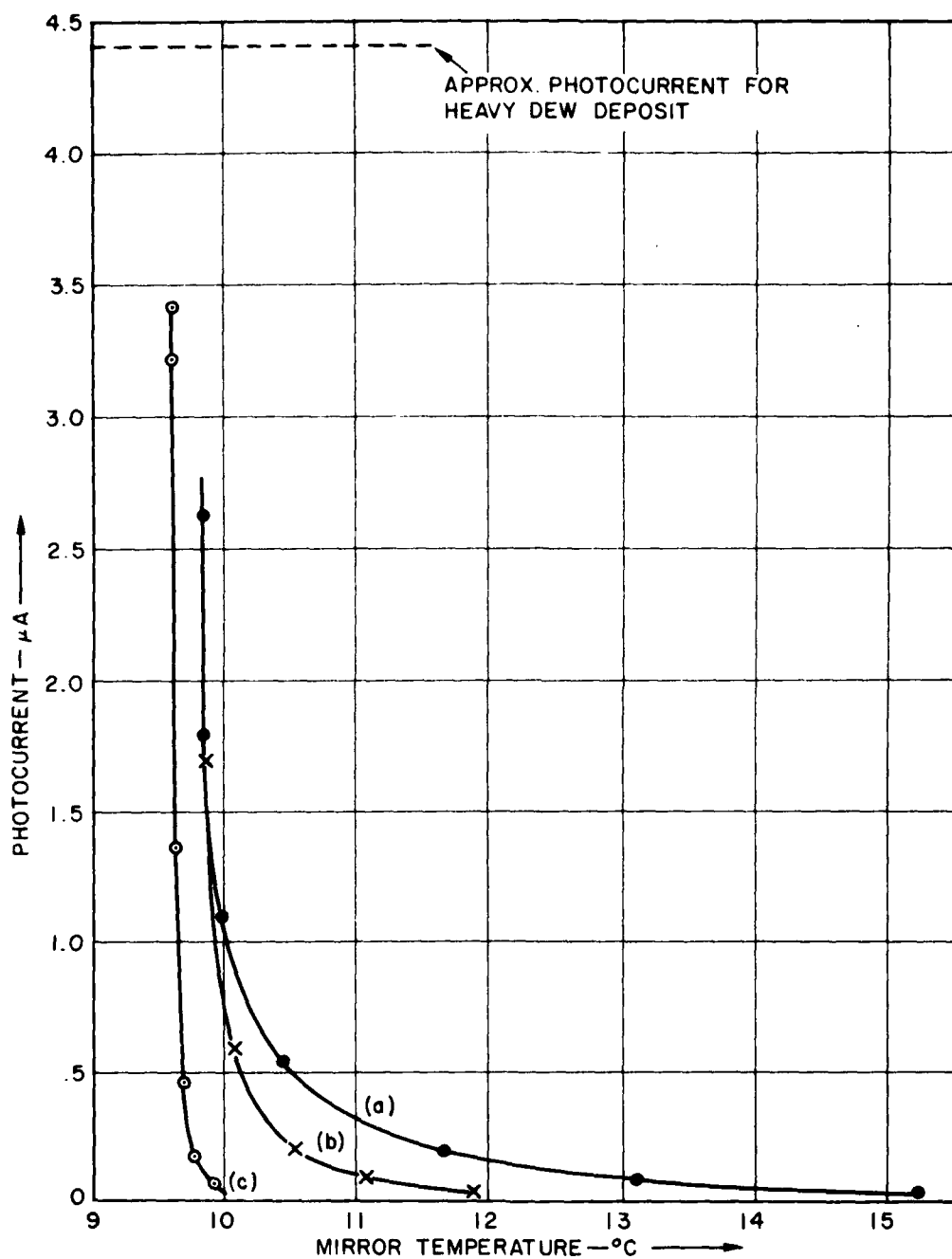


Fig. 5 — Effect of adsorption on dew point which shows dependence of visible scattered light on relative humidity in a photoelectric dew point device. (Martin)

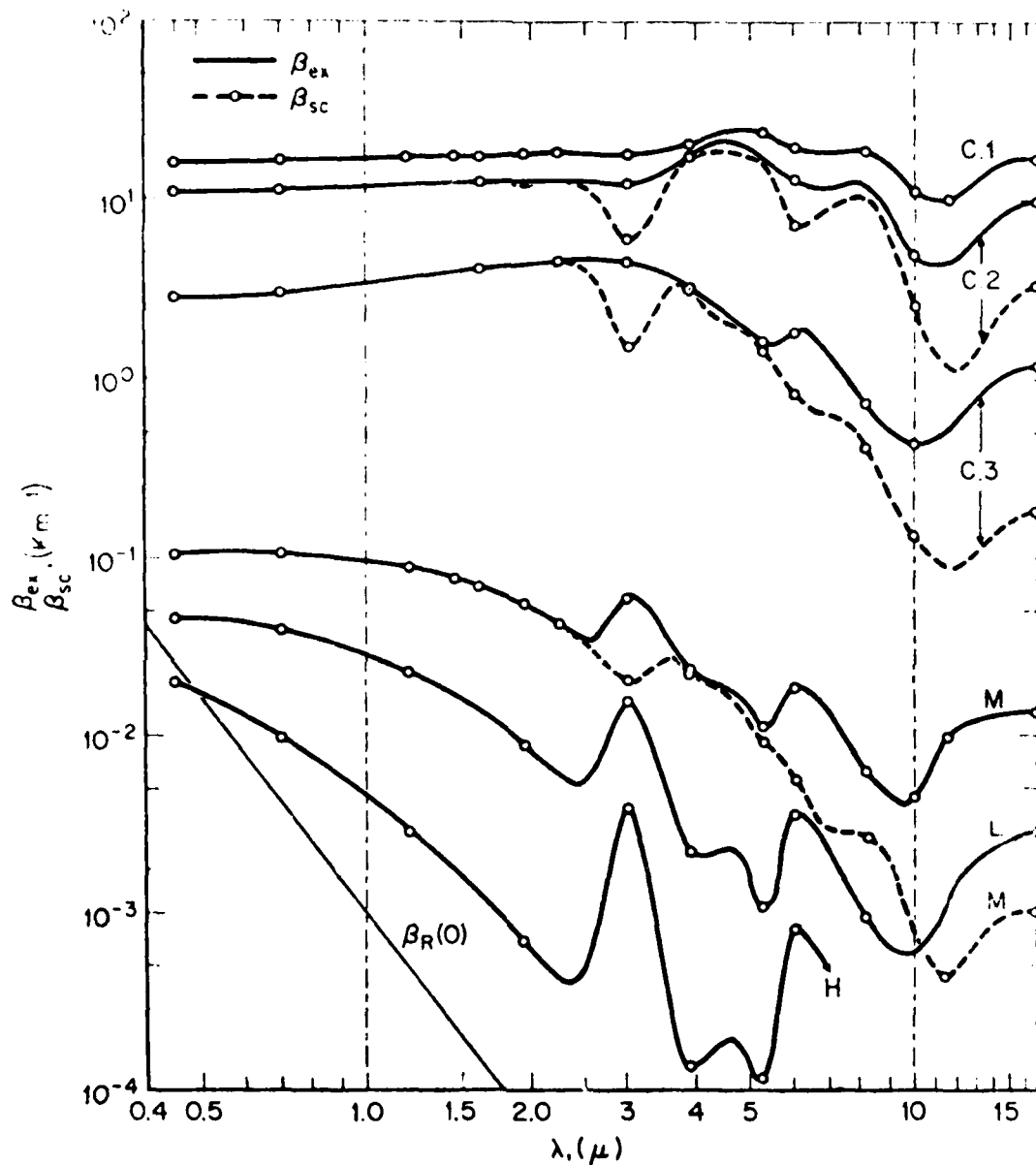


Fig. 6 - Wavelength dependence of the volume extinction coefficient (solid line) and in some cases, of the scattering coefficient (dashed line) for various distributions of water spheres equivalent to 100 particles per cm^3 in each case. (Diermendjian)